REMARKS

A new specification is enclosed which contains all the changes as indicated in this preliminary amendment. All of the amendments involve the re-arranging of subject matter contained in the PCT specification as originally filed, and for clarification, without introducing any new matter. For example, new paragraph 30 correcponds to the content of original Figure 10.

In the Claims, amendments have been made to avoid multiple dependent claims.

Figure 1 has been amended to agree with Scheme 2 on former original page 15, now page 18.

Figure 22 has been deleted since it has been determined that Figure 22 is a duplicate of Figure 8.

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AMENDMENTS TO THE SPECIFICATION:

Page 1, line 2, add a new first paragraph:

[01] This application is a National Stage application of PCT Application PCT/CA2003/001696 filed 07 November 2003 which claims benefit of US Provisional Application 60/424,699 filed 08 November 2002 and US Provisional Application 60/439,811 filed 14 January 2003.

Page 1, Paragraph 4

[04] In the fabrication of organic or polymeric light emitting devices it is advantageous to incorporate multi-layered structured materials with special functions into different location within the device. For instance, organic or polymer light-emitting diodes (LEDs) with a stack of hole-transporting, electron-transporting, and light-emitting layers exhibit enhanced device efficiency, higher brightness, and better stability. Solvent-based or wet-processing techniques such as reel-to-reel printing, screen-printing or spin-coating are important fabrication techniques that could significantly reduce fabrication costs of organic/polymer devices. However, the fabrication of multi-layer device structure is often difficult with wet-processing techniques. One typical problem of making multi-layered structures using solutions is the fact that the solvent used for each successive ~~layers~~ <u>layer</u> can lead to swelling or dissolution of underlying layers.

Page 4, Add new Paragraph 22

[022] According to an embodiment of this aspect of the invention, the boronic acid or boronic acid derivative is selected from the group consisting of a) a single boronic acid, b) a composite of two or more different boronic acids, c) a composite of one boronic acid or its derivative with one or more other materials which contain groups reactive with boronic acid or its derivative, and d) a composite of any number of boronic acids or its derivatives (at least one) with any number and any kinds of other materials which

contain groups listed in the formulas at Figure 10, reactive with boronic acid or its derivative.

Page 5, add new paragraph 24

[024] According to an embodiment of this aspect of the invention, the boronic acid or boronic acid derivative is selected from the group consisting of a) a single boronic acid, b) a composite of two or more different boronic acids, c) a composite of one boronic acid or its derivative with one or more other materials which contain groups reactive with boronic acid or its derivative, and d) a composite of any number of boronic acids or its derivatives (at least one) with any number and any kinds of other materials which contain groups listed in the formulas at Figure 10, reactive with boronic acid or its derivative.

Page 5, paragraph 23 has not become paragraph 25 and has been amended as follows:

[025] According to a yet further aspect of the invention, a multi-layer material is provided, comprising a plurality of layers of a cross-linked composite of boronic acid or a boronic acid derivative such as a boronate, and an organic or organo-metallic moiety having a functionality such as hole transporting, electron transporting and light emitting, wherein each layer has a different functionality.

Page 5, add new paragraph 26:

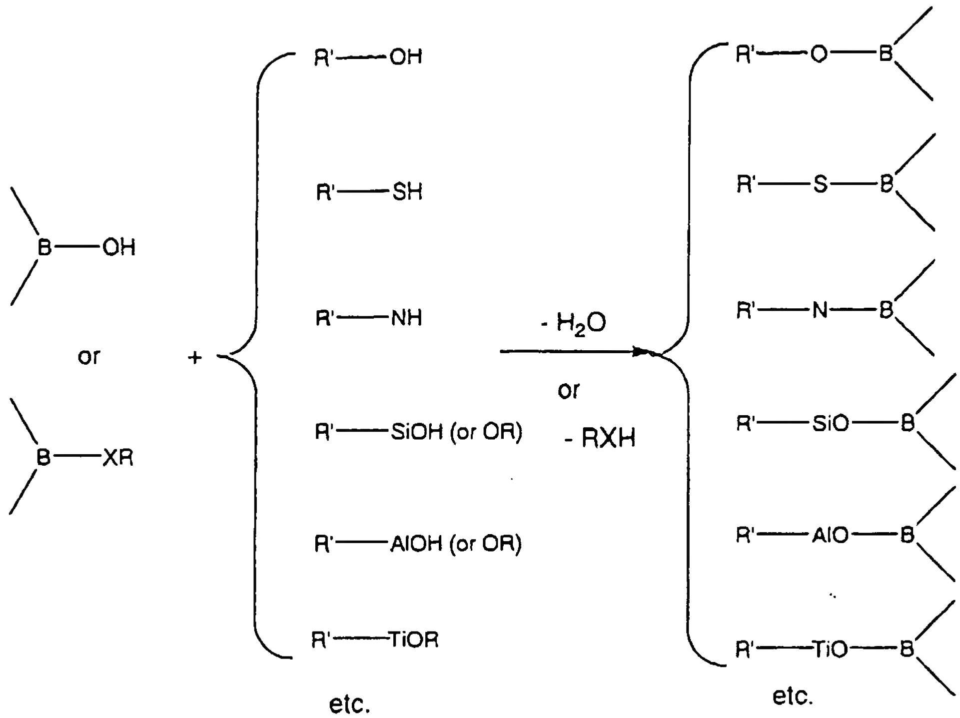
[026] According to a further aspect of the invention, a method is provided for making a multi-layer material comprising a plurality of layers of a cross-linked composite of boronic acid or a boronic acid derivative such as a boronate, and an organic or organo-metallic moiety having a functionality such as hole transporting, electron transporting and light emitting, the method comprising forming on a substrate, a layer of a composite of boronic acid or a boronic acid derivative and an organic or organo-metallic moiety having a functionality such as hole transporting, electron transporting and light emitting, and cross-linking, and forming at least one another such layer having a different functionality such as hole transporting, electron transporting and light emitting and cross-linking.

Page 5, paragraph 24 is now paragraph 27 and has been amended as follows:

[027] According to a further aspect of the invention, a multi-layer optoelectronic device, such as an OLED, is provided, comprising in sequence, a transparent substrate layer, a transparent electrode layer, a layer of a cross-linked composite of boronic acid or a boronic acid derivative such as a boronate, and an organic or organo-metallic moiety having a functionality such as hole transporting, electron transporting and light emitting, at least one another such layer having a different functionality such as hole transporting, electron transporting and light emitting, and another transparent electrode layer.

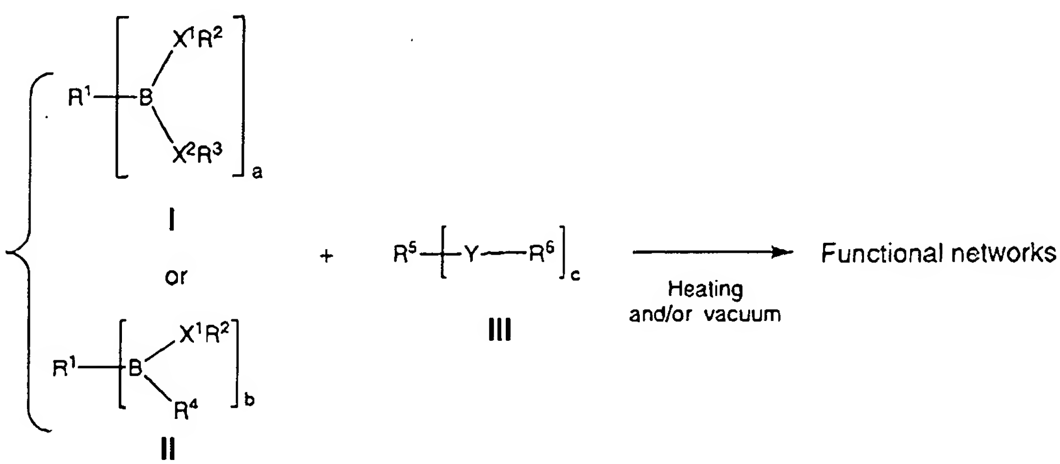
Page 6, add new paragraph 30:

[030] According to yet another aspect of the invention a method of making cross-linked functional networks is provided, comprising reacting compound of structural formula I or structural formula II with a compound of structural formula III(as shown in figure 10), and cross-linking



Equation (1)

wherein, X = O, S, N; R and R' = alkyl, aryl, or any other organic and inorganic structures or groups.



Equation (2)

wherein,

R^1, R^4, and R^5 = alkyl, aryl, or other groups, either organic or inorganic, but at least one of them contains functionality; they can be of small molecular weights or high molecular weights.

R^2, R^3, R^6 = H, alkyl, aryl, they may be same or different, but at least one of them is H.

X^1, X^2 = O, S, or N, they may be same or different.

Y = O, S, N (or NH), SiO, AlO, TiO, etc.

a,b, c are equal to or larger than one, but at least either a (or b) or c is larger than one.

Page 6, add new paragraph 31:

[031] According to yet another aspect of the invention a cross-linked functional network is provided, made by the method described in the preceding paragraph.

Page 6, paragraph 31 has now become paragraph 35 and has been amended as follows:

[035] Figure 4 is a schematic illustration of ~~three~~ two single layer(b and c) and one double layer(a) structures according to the invention.

Page 7, delete paragraphs 46 and 47 and add the following paragragh:

[050] Figure 22 is a graph illustrating the EL emission at different driving voltages.

Page 8, paragraph 50 has now become paragraph 52 and has been amended as follows:

[052] In this invention, we demonstrated the use of organoboronic acids as cross-linking end groups (or side groups) to immobilize the functional materials in a thin film for multi-layer device fabrication. . A double layer organic light emitting diode from CzBA and F_3BA ~~(Figures 2 and 3)~~ (Figure 4(a)) has been fabricated as a testing device to demonstrate that a multi-layered device structure can be easily realized using materials containing organoboronic acid groups. Since the OLED shown here was only used as a demonstration, we expect this film processing technique (using organoboronic acid groups as end caps) can be applied to the fabrication of photovoltaic cells, photonic multi-layer structures, and any other organic electronic/optical devices which require a multi-layered structure.

Page 9, Paragraph 60 has now become paragraph 62 and has been amended as follows:

[062] The ~~advantange~~ advantage of the double layer structure:

Page 9, paragraph 61 has now become paragraph 63 and has been amended as follows:

[063] For the comparison, three device ~~streteures~~ structures were used as seen in Figure 4.

Page 10, paragraph 62 has now become paragraph 64 and has been amended as follows:

[064] Devices with a double layer structure were successfully fabricated using CzBA and F_3BA (Figure 4(a)). The CzBA layer functioned as a hole-transporting layer, and F_3BA functioned as an emissive and electron-transporting layer. The following are the device performance data of OLEDs based on a CzBA and F_3BA double layer structure, and F_3BA and CzBA single layer structures:

Page 10, paragraph 63 has now become 65 and has been amended as follows:

[065] 1. At the same electric field strength, the double layer structure device conducts much more electric current (current =hole current + electron current) than the single layer devices (Figure 5).

Page 10, paragraph 67 has now become 69 and has been amended as follows:

[069] A double layer structure has been fabricated successfully by spin-coating CzBA and F_3BA solution and the subsequent mild thermal treatment. This demonstrated that the thermal dehydration of diboronic acids to form cross-linked networks could be used as an easy and effective approach to fabricate double or multi-layered devices. Our preliminary experiments on thin film and device processing show that the thin films are quite uniform as indicated by the uniform thickness interference pattern, and uniform light emission area, which depends strongly on the layer thickness. The CzBA layer plays a role of hole-transporting layer, while the F3BA layer functions as both an electron-transporting layer and a light-emitting layer. The fact that the fluorescence intensity did not decrease after a thermal treatment at 130 °C indicates that the cross-linking process is useful in enhancing the thermal stability, especially for fluorene-based compounds, which are know spectrally unstable upon heat treatment. The maximum luminance value recorded for the CzBA/F3BA double layer device is around 170 cd/m^2. This value is obtained from a luminance meter which is calibrated as a photopic response device (Figure 9).

Page 11, paragraph 70 has now become 71 and has been amended as follows:

[071] II. The crosslinking reactions do not require any initiators are can be carried out under quite low temperatures (as low as room temperature) with water as

the only by-product that can be easily removed under vacuum. Therefore there is no generation of highly reactive radicals involved and no harmful initiator redidues residues left in the materials.

Page 12, paragraph 76 has now become 78 and has been amended as follows:

[078] VIII. Because organic light emitting devices are very sensitive to the charge mobility, HOMO-LUMO level positions, and interface quality, the demonstration of a working OLED device ~~indicate~~ indicates a reasonably good interface structure between the CzBA and F3BA layers. Since OLED is only used here as a testing vehicle, we expect the same film processing technique (using organoboronic acid groups as end caps) can be applied to the fabrication of photovoltaic cells, photonic multi-layer structures, and other electronic/optical devices.

Page 16, paragraphs 83 and 84 have now become 85 and 86 and have been amended as follows:

[083] The crosslinkable components containing CzBA/PSA, FnBA/PSA, and FnBOeEt/PSA have been used to form crosslinked hole-transporting and light-emitting thin films to demonstrate the feasibility of this approach (Scheme 1 – 3).

[084] The organoboronic acids, CzBA, FnBA, and poly(styrene-co-allyl alcohol) (PSA) are readily soluble in THF. The THF solutions of CzBA/PSA, FnBA/PSA, and FnBOeEt/PSA can be used to fabricate thin films using traditional approaches such as spin-coating, solution-casting, or dip-coating techniques. Once the film has been prepared it can be crosslinked using mild reaction conditions (e.g., 120 °C, 2h) in which water is the by-product, that can be removed by vacuum (this reaction has been confirmed by FT-IR analysis). This thermal treatment procedure produces crosslinked thin films that are insoluble in water alone or in any anhydrous organic solvents. These films are also resistant to most wet common solvents such as THF and acetone. The crosslinked films produced as outlined above have higher optical stabilities (UV absorption and fluorescence) at elevated temperatures (e.g., 120 °C, 24 h) than the crosslinked structures made solely from boronic acids (see Appendix I below).

Page 31, paragraph 136 and 137 have become paragraphs 138 and 139 and have been amended as follows:

[0138] The electroluminescence spectrum of the CzBA/F_3BA double layer device is identical to the photoluminescence spectrum of F_3BA thin film (peaked ~~@~~ at 416nm). However, the EL spectrum of a single layer F_3BA device is very different from the PL of F_3BA thin film, there is a very broad long-wavelength shoulder beside the fluorene emission peak (See Figure ~~224~~ 8), that might be caused by imbalanced charge injection (electrons >> holes) in the device. In many cases, molecules are much less stable at charged state.

[0139] This series of experiments demonstrated that a double layer structure has been fabricated successfully using CzBA and F_3BA. The hole-transporting layer and electron-transporting layer functioned properly according to the design. That the fluorescence intensity did not decrease after a thermal treatment at 130 °C indicate the cross-link process is useful in enhancing the thermal stability, especially for fluorene-based compounds, which are know thermally unstable. The peak position of the EL emission at different driving voltages remained at 426 nm (See Figure ~~23~~ 22). This is a significant improvement as compared to other polyfluorene based LEDs.

AMENDMENTS TO THE CLAIMS

1. (Previously presented) A cross-linked composite of boronic acid or a boronic acid derivative such as a boronate, and an organic or organo-metallic moiety having a functionality such as hole transporting, electron transporting and light emitting.

2. (Previously presented) A cross-linked composite according to Claim 1, wherein the boronic acid or boronic acid derivative is selected from the group consisting of a) a single boronic acid, b) a composite of two or more different boronic acids, c) a composite of one boronic acid or its derivative with one or more other materials which contain groups reactive with boronic acid or its derivative, and d) a composite of any number of boronic acids or its derivatives (at least one) with any number and any kinds of other materials which contain groups listed in the formulas at Figure 10, reactive with boronic acid or its derivative.

3. (Currently amended) A cross-linked composite according to Claim 1 ~~or 2~~, wherein the organic or organometallic moiety is a mixture of such moieties having a different functionality.

4. (Previously presented) A method of making a cross-linked composite of boronic acid or a boronic acid derivative such as a boronate, and an organic or organo-metallic moiety having a functionality such as hole transporting, electron transporting and light emitting, comprising attaching to boronic acid or a boronic acid derivative, an organic or organo-metallic moiety having a functionality such as hole transporting, electron transporting and light emitting, and cross-linking.

5. (Previously presented) A method according to Claim 4, wherein the boronic acid or boronic acid derivative is selected from the group consisting of a) a single boronic acid, b) a composite of two or more different boronic acids, c) a composite of one boronic acid or its derivative with one or more other materials which contain groups reactive with boronic acid or its derivative, and d) a composite of any number of boronic acids or its derivatives (at least one) with any number and any kinds of other materials

which contain groups listed in the formulas at Figure 10, reactive with boronic acid or its derivative.

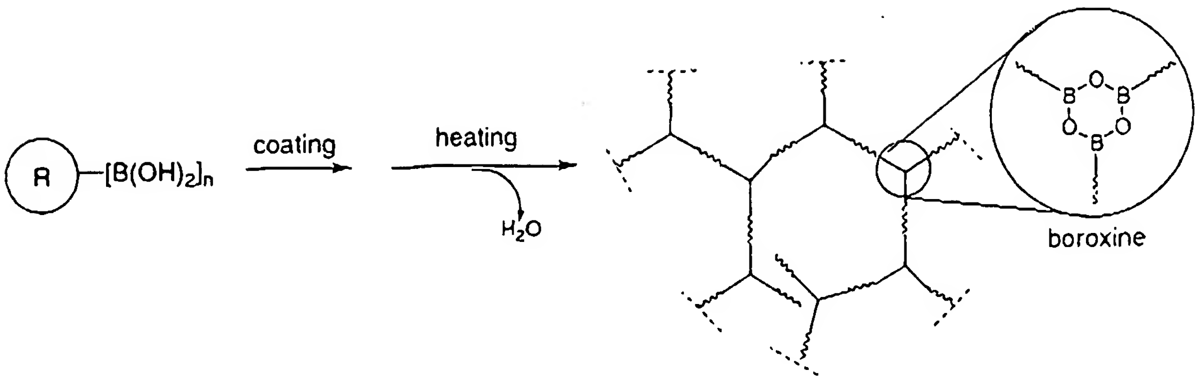
6. (Currently amended) A method according to Claim 4 ~~or 5~~, wherein the organic or organometallic moiety is a mixture of such moieties having a different functionality.

7. (Previously presented) A multi-layer material comprising a plurality of layers of a cross-linked composite of boronic acid or a boronic acid derivative such as a boronate, and an organic or organo-metallic moiety having a functionality such as hole transporting, electron transporting and light emitting, wherein each layer has a different functionality.

8. (Previously presented) A multi-layer material according to Claim 7, wherein the boronic acid or boronic acid derivative is selected from the group consisting of a) a single boronic acid, b) a composite of two or more different boronic acids, c) a composite of one boronic acid or its derivative with one or more other materials which contain groups reactive with boronic acid or its derivative, and d) a composite of any number of boronic acids or its derivatives (at least one) with any number and any kinds of other materials which contain groups listed in the formulas at Figure 10, reactive with boronic acid or its derivative.

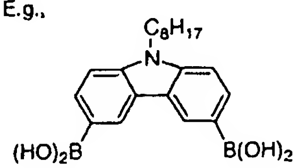
9. (Currently amended) A multi-layer material according to Claim 7 ~~or 8~~, having two functional layers, one functional layer having a hole transporting functionality, and the other functional layer having a a light emitting functionality and an electron transporting functionality.

10. (Previously presented) A multi-layer material according to Claim 9, wherein the layer having the hole transporting functionality comprises CzBA, and wherein the layer having the light emitting functionality and an electron transporting functionality is F_nBA, wherein n= 2, 3 or 4.

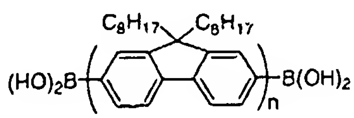


n = or >1;
R = organic or organometallic complex moiety including oligomer and polymer.

E.g.,

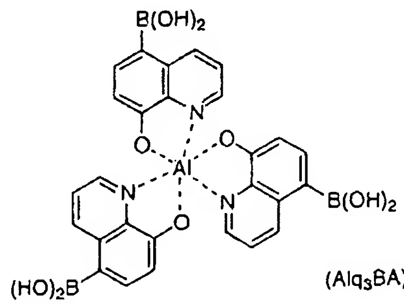


(CzBA)



~~n = 1, 2, 3, and 4~~
n = 2, 3 and

(F_nBA)



(Alq_3BA)

Fig. 1